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Mechanoluminescence produced during impulsive deformation of coloured alkali halide crystals

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Abstract Mechanoluminescence (ML) in coloured alkali halide crystals is reported. During impulsive excitation of γ-irradiated alkali halide crystals, two peaks are observed in the ML intensity *versus* time curves. The first peak lies in the deformation region and the second peak in the post-deformation region. The ML in the deformation region is due to the recombination of dislocation-trapped electrons with the holes in defect centres. The ML in the post-deformation region is due to the transient thermostimulated luminescence from shallow traps which get populated due to the Auger process occurring during transfer of dislocation-trapped electrons to deep traps. In the frame work of the mechanism proposed, an equation is derived which is able to explain the ML in the deformation and post-deformation region of the crystal. Several other facts related to the ML produced during impulsive excitation of coloured alkali halide crystals, are also discussed.

Keywords Mechanoluminescence, dislocations, deformation, alkali halides

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1. Introduction

Mechanoluminescence (ML), the phenomenon of cold light emission induced during mechanical deformation of solids, links the mechanical, spectroscopic, electrical and structural properties of solids. A large number of organic and inorganic solids exhibit the phenomenon of ML. The ML of coloured alkali halide is considered to be especially interesting due to the following reasons : (i) the ML in these crystals appears not only during their fracture, but also during their plastic and elastic deformation, (ii) the ML has memory effect *i.e.* if a crystal is deformed upto a particular stress, then it shows the ML in the next deformation cycle only after the deformation occurred in the previous cycle, and (iii) the ML intensity depends linearly on the density of colour centres in the crystals [1-10].

Most of the crystals exhibit ML only during their deformation and ML emission stops as soon as the deformation is interrupted. When such crystals are deformed mechanically the ML intensity initially increases, attains a maximum value and then decreases. Thus, one peak is observed in a plot between the ML intensity and time.

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However, when the ML is excited impulsively in γ-irradiated alkali halide crystals, the ML appears in the deformation region as well as in the post-deformation region. Two peaks are observed in the ML intensity *versus* time curve where the first peak lies in the deformation region but the second peak lies in the post-deformation region. The appearance of ML in the post-deformation region and occurrence of two peaks in the ML *versus* time curves of coloured alkali halide crystals are not yet understood. The present paper reports the ML produced during impulsive deformation of γ -irradiated crystals of NaCl, NaBr, KCl, KBr and KI.

2. Experimental

The experimental arrangement for impulsive excitation of ML in γ-irradiated alkali halide crystals was described previously [11]. For measuring the ML activity, crystal was placed on a transparent lucite plate below the guiding cylinder. An RCA 931 photomultiplier tube was used for monitoring the luminescence from below the lucite plate. The crystal was covered with a thin aluminum foil and fixed with an adhesive tape. The ML was excited impulsively by dropping a hollow cylinder of 800g (2cm dia) from different height through a guiding hollow cylinder.

The output of the PMT was connected to a scientific HM 307 oscilloscope having P7 phosphorescent screen capable of sustaining a trace in dark for more than a minute. The ML *versus* time curve was determined by recording the trace onto the oscilloscope screen and total ML intensity was determined by measuring the area below this curve.

3. Results

When a piston is dropped on to γ -irradiated crystals of NaCl, NaBr, KCl, KBr and KI, the ML emission is observed.

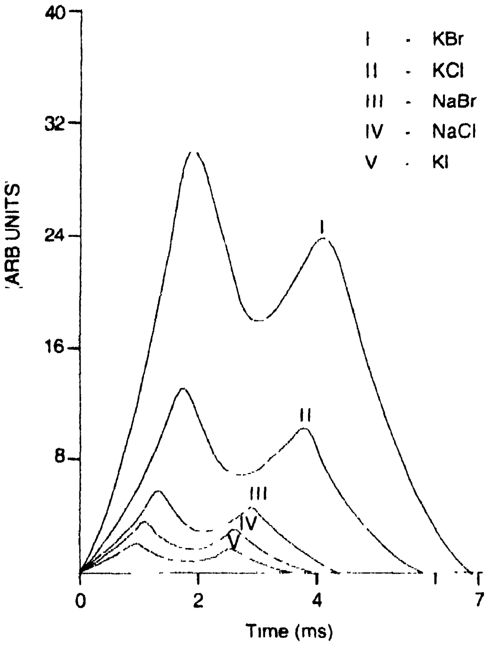

Figure 1. Time dependence of the ML intensity of γ-irradiated crystals for different materials.

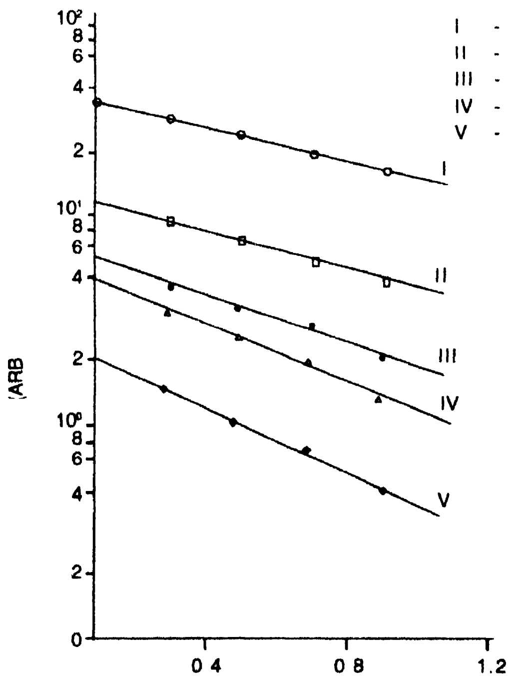

Figure 2. Plot of log I *versus* $(t-t_{m})$ of γ-irradiated crystal of different materials.

Figure 1 shows the time-dependence of the ML intensity of γ -irradiated crystals of different materials. The ML emission in these crystals is found in deformation as well as post-deformation region. It is seen that I_{m1}, I_{m2}, t_{m1}, and t_{m2} increase with increasing dimensions of the crystals. Figure 2 shows the plot of log **I** *versus* $(t-t_{m1})$ of γ - irradiated crystals for different

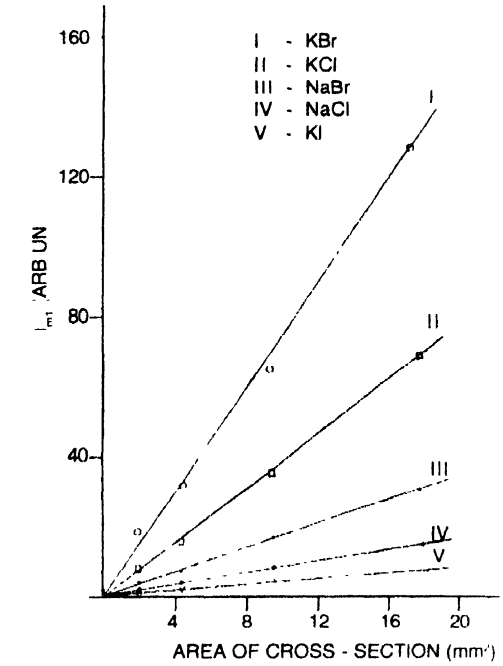

Figure 3. Dependence of the peak ML intensity I_{m1} on the area of cross section for different materials

materials. It is seen that the plot is a straight line with negative slope, the slope being different for different materials. Figure 3 shows the plot of I_{m1} *versus* area of cross section for different

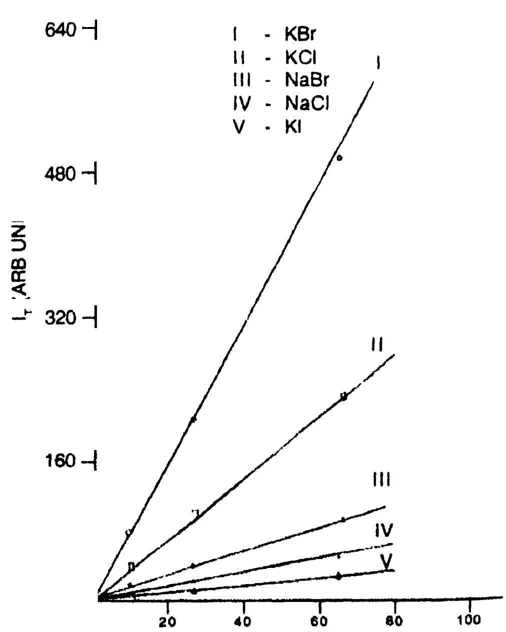

Figure 4. Dependence of the total ML intensity I_T on the volume of the crystals for different materials.

materials. It is found that I_{m1} increases linearly with areas of cross sections of the crystals. Figure 4 shows the plot of I_t *versus* volume of different crystals. It is found that I_t increases with increasing volumes of the crystals. Figure 5 show the plot of I_{m1} *versus* γ -dose for different materials. It is seen that initially I_{m1} increases with γ -dose and then it tend to attain a saturation value. Figure 6 shows the plot of total ML intensity I_t *versus* γ -dose for different crystals. It is seen that initially I_t increases with γ - dose and then tends to attain saturation value.

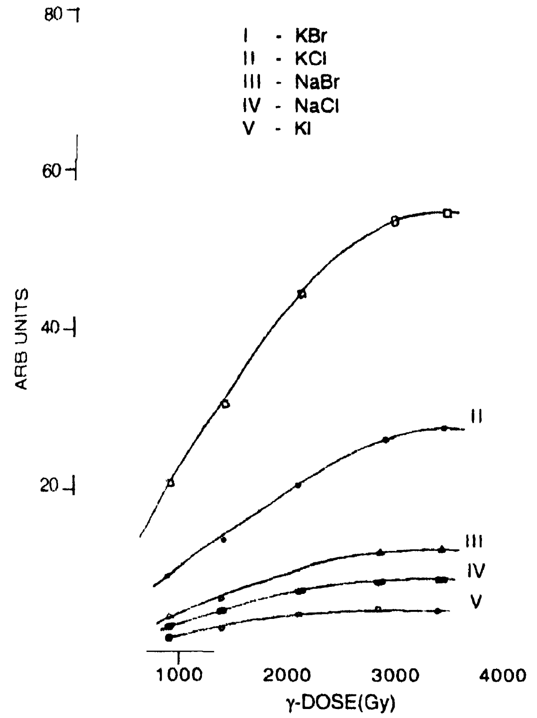

Figure 5. Dependence of the peak ML intensity I_{m1} on γ-irradiation dose for different materials

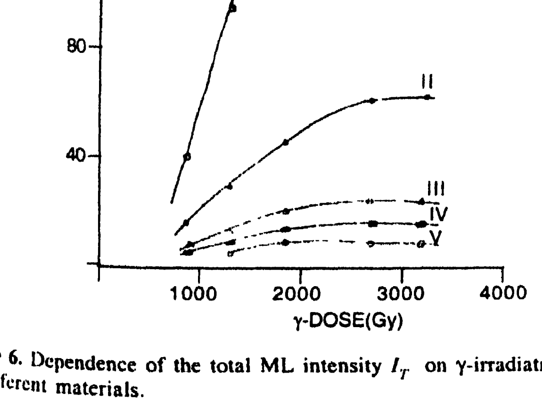

Figure 6. Dependence of the total ML intensity I_T on γ-irradiation dose for different materials.

4. Discussion

When a piston impacts on to a crystal with an initial velocity v_o, then its velocity v at any time t may be expressed by the relation

$$dv/dt = -\beta v\,, \tag{1}$$

where β is attrition co-efficient.

If λ_d is the mean free path of the dislocations, r_i is the radius of interaction between the moving dislocations and F-centre, n_i is the density of F-centres, then the number of F-centre electrons interacting with one dislocation will be $\lambda_d\, r_F\, n_F$. If V is the volume of the crystal, b is the Burger's vector and H is the thickness of the crystal, then the number of F-centre electrons interacting with dislocation per second may be written as

$$g_i = \left\{\lambda_d\, r_F\, n_F\, v_0\, V/bH\right\} \exp(-\beta t)\,. \tag{2}$$

In the expansion region of edge dislocation, the average ground state energy E_i of the F-centre interacting with dislocation is higher as compared to the non-interacting F-centre [12]. Thus, E_i will be in between the normal ground state of F-centre and the dislocation band.

If α_1 is the rate constant for jumping of F-centre electrons to the dislocation band and α_2 is the rate constant for the dropping back of the interacting F-centre electrons to the normal F-level, then

$$\alpha = \alpha_1 + \alpha_2\,.$$

If P_i is the probability of capture of F-centre electrons by the moving dislocations, then the rate of generation of electrons in the dislocation band may be given by

$$g = \left\{\lambda_d\, p_F r_I\, n_F v_0\, V/bH(\alpha-\beta)\right\}\left[\exp(-\beta t) - \exp(-\alpha t)\right]. \tag{3}$$

The electrons moving with dislocations may recombine with holes. They may also be captured by deep traps and stationary dislocation states and other compatible traps. If $\sigma_1, \sigma_2, \sigma_3$ and σ_4 are the crosssections and N_1, N_2, N_3 and N_4 are the densities of holes, deep-traps, stationary dislocation states and other compatible traps, respectively.

If $\mathcal{Y} = 1/\tau_d = (\sigma_1 N_1 + \sigma_2 N_2 + \sigma_3 N_3 + \sigma_4 N_4)\, v_d$ is the lifetime of the electrons captured by moving dislocations, then the number of dislocation electrons at any time t may be given by

$$n_d = \left\{\lambda_d\, p_F r_F\, n_F v_0\, V/bH\mathcal{Y}(\alpha-\beta)\right\}\left[\exp(-\beta t) - \exp(-\alpha t)\right]. \tag{4}$$

The rate of recombination electrons moving with dislocations with the hole centres may be given by

$$R_d = \sigma_1 N_1 v_d n_d\,.$$

If η is the probability of radiative recombination of the electrons moving with the dislocations with the hole centres then the ML intensity may be given by

$$I_1 = \eta\, \sigma_1 N_1 v_d \lambda_d\, p_F r_F\, n_F v_0\, V/bH\gamma(\alpha - \beta)$$
$$\times\left[\exp(-\beta t) - \exp(-\alpha t)\right]$$

or

$$I_1 = \left\{\eta\sigma_1 N_1 \lambda_d p_F r_F n_F v_0\, V/bH\left(\sigma_1 N_1 + \sigma_2 N_2 + \sigma_3 N_3 + \sigma_4 N_4\right)\right.$$
$$\left.\times(\alpha - \beta)\right\}\left[\exp(-\beta t) - \exp(-\alpha t)\right]$$

or $$I_1 = I_0\left[\exp(-\beta t) - \exp(-\alpha t)\right], \tag{5}$$

where $I_0 = \eta\, \sigma_1 N_1\, \lambda_d p_F r_F\, n_F v_0\, V/bH$

$$\times\left(\sigma_1 N_1 + \sigma_2 N_2 + \sigma_3 N_3 + \sigma_4 N_4\right)(\alpha - \beta).$$

It is to noted that in the dislocation energy band, an electron participates in two motions. It may travel along a dislocation (because the dislocation band is one dimensional) and it can travel with the dislocation. As the electrons move with very low velocity along the dislocation, the lifetime, which depends. As the electrons move with very low velocity along the dislocation, the lifetime which depends inversely on the electron velocity may be long in the stationary dislocations as compared to that in the moving dislocations [7,12].

The rate of transfer of electrons from moving dislocation to the stationary dislocation is given by

$$g_s = \sigma_3 N_3 v_d n_d.$$

If $\tau_s = 1/\mathcal{Y}_s$ is the lifetime of electrons in the stationary dislocations, then the ML intensity produced due to the motion of electrons in the band of stationary dislocations may be written as

$$I_2 = \left\{\eta \mathcal{Y}_s \sigma_3 N_3\, \lambda_d p_F r_F n_F v_0\, V/bH\right.$$
$$\left.\times\alpha\left(\sigma_1 N_1 + \sigma_2 N_2 + \sigma_3 N_3 + \sigma_4 N_4\right)(\beta - \mathcal{Y}_s)\right\}$$
$$\times\left[\exp(-\mathcal{Y}_s t) - \exp(-\beta t)\right]$$

or $$I_2 = I_0''\left[\exp(-\mathcal{Y}_s t) - \exp(-\beta t)\right], \tag{6}$$

where

$$I_0'' = \eta \mathcal{Y}_s \sigma_3 N_3\, \lambda_d p_F r_F n_F v_0\, V/bH\alpha$$
$$\times\left(\sigma_1 N_1 + \sigma_2 N_2 + \sigma_3 N_3 + \sigma_4 N_4\right)(\beta - \mathcal{Y}_s).$$

It is to be noted that in the case of fast deformation, the diffusion of holes towards the F-centres, as well as the recombination of dislocation electrons with holes may also give rise to ML. But the intensities of ML produced due to these processes may be negligible as compared to I_{m1} and I_{m2} and therefore, they would not be detected during the measurement of the time-dependence of the ML produced during the fast deformation of the crystals.

4.1 Estimation of t_{ml} :

It is seen from eq. (5) that $I_1 = 0$, for $t = 0$ and $t = \infty$, thus the ML intensity should be maximum for a particular value of time. For maximum value of I_1, dI_1/dt should be equal to zero. Differentiating eq. (5) and equating dI_1/dt to zero, and writing the time corresponding to the first peak in the ML intensity *versus* time curve as t_{m1}, we get

$$t_{ml} = 1/(\alpha - \beta)\ln(\alpha/\beta) \approx 1/\alpha\ln(\alpha/\beta).$$

As β increases with the strain rate and α is independent of the strain rate, it seems that t_{m1}, should decrease slowly with the impact velocity v_0 of the hammer.

4.2 Estimation of I_{ml} :

Substituting the value of t_{m1} from eq. (7) in eq. (5), we get the ML intensity corresponding to the first peak in the ML intensity *versus* time curve as

$$I_{m1} = I_0',$$
$$I_{m1} = \eta\, \sigma_1 N_1\, \lambda_d p_F r_F\, n_F v_0\, V/bH\alpha$$
$$\times\left(\sigma_1 N_1 + \sigma_2 N_2 + \sigma_3 N_3 + \sigma_4 N_4\right)$$

4.3 Estimation of t_{m2} :

From eq. (6), we get one value of the time corresponding to the second peak in the ML intensity *versus* time curve as

$$t_{m2} = 1/(\beta - \mathcal{Y})\ln\beta/\mathcal{Y}_s \approx 1/\beta\ln(\beta/\mathcal{Y}_s).$$

4.4 Estimation of I_{m2} :

Substituting the value of t_{m2} from eq. (9) in eq (6), we get the ML intensity corresponding of the second peak in the ML intensity *versus* time curve as

$$I_{m2} = I_0'',$$
$$I_{m2} = \eta \mathcal{Y}_s \sigma_3 N_3 \lambda_d p_F r_F n_F v_0\, V/bH\alpha$$
$$\times\left(\sigma_1 N_1 + \sigma_2 N_2 + \sigma_3 N_3 + \sigma_4 N_4\right)(\beta - \mathcal{Y}_s).$$

4.5 Total ML intensity I_t :

The total ML intensity is given by

$$I_T = \int_0^\infty I\, dt.$$

From eqs. (5), (6) and (11), we get

$$I_T = \int_0^\infty I_0'\left[\exp(-\beta t) - \exp(-\alpha t)\right]dt$$
$$+ \int_0^\infty I_0''\left[\exp(-\mathcal{Y}_s t) - \exp(-\beta t)\right]dt,$$

$$I_T = \{\eta(\sigma_1 N_1 + \sigma_3 N_3)\lambda_d p_F r_F n_F v_0 \, V/bH\alpha\beta$$
$$\times(\sigma_1 N_1 + \sigma_2 N_2 + \sigma_3 N_3 + \sigma_4 N_4). \qquad (12)$$

5. Conclusions

(i) During the impulsive deformation of γ-irradiated crystals of NaCl, NaBr, KCl, KBr and KI, two peaks are observed in the ML intensity *versus* time curve. The first peak I_{m1} which appears in the deformation region, is always greater then the second peak I_{m2} which appears in the post-deformation region. The ML intensity after peak I and peak II decays exponentially with time.

(ii) For the impulsive excitation of ML in coloured alkali halide crystals, the values of peak ML intensity I_{m1} and the total ML intensity I_T increases linearly with the area of cross section of the crystals while the peak ML intensity I_{m2} varies linearly with the volume of the crystals. The values of I_{m1}, I_{m2} and I_T are different for different materials.

(iii) The time t_{m1} corresponding to the first peak I_{m1} increases slowly and the time t_{m2} corresponding to the second peak I_{m2} increases significantly with increasing thickness of the crystals, the time t_{m1} and t_{m2} are different for different crystals for impulsive deformation of coloured alkali halide crystals.

(iv) For impulsive excitation, I_{m1}, I_{m2} and I_T initially increase with γ-dose and finally attain saturation values for higher values of γ-dose given to the crystals. The time t_{m1} and t_{m2} do not change significantly with change in irradiation dose given to the crystals.

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